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TREATMENT AND DISPOSAL OF ATOMIC ENERGY WASTE CONTAINING RADIOACTIVE ISOTOPES*

Mr. Hayner's paper has covered the policy of the Atomic Energy Commission with respect to many problems on the treatment and disposal of high level radioactive wastes. Gorman (1) has presented a discussion of the sanitary engineering approach to the waste disposal problems of the industry. He points out that the effectiveness and economy of the recommended procedures have found wide acceptance by American industrial management and refers to research in progress supported by the Atomic Energy Commission.

It is probable that no new industry has given such serious consideration to its waste disposal problems in the interest of the public welfare than has the American atomic energy industry. Realization within this industry of the environmental aspects of waste disposal as well as its economic significance is a most important consideration and is setting a good example for all new industries.

In sharp contrast with wastes of other industries, radioactive wastes may not be objectionable as measured by such common characteristics as odors, tastes and biochemical oxygen demand, yet they may be most dangerous because of levels of radioactive energy and long periods of activity. Furthermore, their cumulative effects on living organisms are not too well understood.

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The intricate and carefully controlled processing of radioisotopes and their manyfold uses in production and research ultimately lead to radioactive wastes in solid, liquid and gaseous forms; and treatment of a waste for decontamination in one form may result in radioactive wastes in another. For example, disposal of combustible solids by incineration create radioactive gases, and treatment of radioactive solutions by evaporation results in concentrates in the form of sludges or dry solids. In all cases an ultimate problem of disposal of high or low level wastes must be solved so that the public health is fully protected while the cost of operation is held to a minimum.

It is pertinent to call attention to the diversity of the radioactive wastes currently being obtained from various segments of this industry and to emphasize that as the industry expands the waste disposal problem may become even more complex. There is a considerable difference in the composition and complexities, the hazards, the kind of radiation and the quantities of wastes arising from mining and refining operations, atomic piles or medical and research laboratories. Although all of these sources contribute wastes, these may vary tremendously in quantities and characteristics and may require different treatment procedures or at present even may be untreatable. There are differences in characteristics, quantities and treatability of atomic reactor wastes depending upon the operating power level, the type of coolant used, the kind of fuel, the period of storage of irradiated material and whether the reactor is operated for plutonium production, power development or production of radioisotopes.

For instance, although waste disposal associated with the new small reactor that is under construction at North Carolina State College will present some problems, they will be minor compared to similar problems at the Oak Ridge National Laboratory. Numerous classified reports dealing with the waste problem of the industry have been prepared. An examination of the abstracts of some 50 unclassified reports on radioactive waste disposal by Shannon [2] will illustrate the diversity of these problems. The review of the unclassified literature by Setter of the Federation of Sewage and Industrial Wastes Associations' Research Committee [3] summarized the development of permissible tolerance levels for radioisotopes in drinking waters and the problems and research programs on waste disposal.

In establishing a perspective of these disposal problems compared to common problems and effluent standards for the disposal of non-radioactive wastes, it is instructive to examine some suggested tolerance values for drinking waters. For example, Morgan's [4] suggested drinking water tolerance values for two radiotoxic isotopes are 0.58 microcuries per liter for Sr^{90}, Y^{90} and 5.4 microcuries per liter for Cs^{137}. In terms of concentration units these limits are equivalent to 0.0035 p.p.b. for Sr^{90}, Y^{90} and 0.065 p.p.b. for Cs^{137}. It is evident that before a process for waste treatment is acceptable it must be proven capable of attaining the very stringent effluent standards. Furthermore, in establishing the degree of waste treatment to

be required of users of radioactive materials, consideration should properly be given to subsequent dilution factors which may be expected after release of the wastes and which could be depended on to lessen the effect of contamination.

Hazardous wastes for which no practical or economical methods for treatment, disposal or recovery are yet available are stored at sites controlled by the Atomic Energy Commission. Storage of high level mixed fission wastes is still a common practice of the industry though studies on chemical processing and evaporation, such as those referred to in the previous paper, may reduce this practice. Considerable emphasis is being given to development of processes for recovery of high level waste for use of the energy contained within them. It is to be expected, however, that at a certain low level of activity these wastes cannot be used economically. Development of low cost and feasible methods of treatment and disposal of these wastes, some of which are quite voluminous is in progress. Such development is patterned after physical chemical and biochemical processes used in disposal of domestic sewage and other ordinary industrial wastes.

The present paper will discuss the application of the common coagulation and carrier precipitation procedures that are common in municipal and industrial water purification to the treatment of liquid radioactive wastes. These procedures are satisfactory for treatment of wastes containing concentrations of some relatively long life isotopes

too low in activity to justify on economic grounds prolonged storage or decontamination by evaporation, but still too high for innocuous discharge into a surface or ground water. Relatively dilute wastes which are produced in considerable volume may be economically treated by adaptation of relatively simple water purification processes.

Experiments at Los Alamos on the treatment of laboratory wastes containing plutonium have been described by Christenson et al (5). These wastes resulted from the activities of research laboratories. Since the research activities of the laboratories are unpredictable, the composition of the wastes cannot be anticipated. It was pointed out that the total solids content of 8 hour composites of this waste varied from 230 to over 8,000 p.p.m. The plutonium activity of the wastes was found to vary from 200 to 25,000 counts per minute per liter. Since 1 microgram of plutonium equals 70,000 counts or at 50 per cent geometry, 140,000 disintegrations per minute it will be noted that the quantities contained in these wastes are very low and rarely approach 1 microgram per liter. These wastes also contain complexing and sequestering agents such as citric acid and polyphosphates which are used in laboratory and apparatus decontaminating processes and which increase the difficulties of treatment. The objective of the treatment is to reduce the plutonium activity to 70 counts per minute per liter or less.

The laboratory and pilot plant experiments showed that although the complexing agents interferred with coagulation procedures, coagulation treatment with iron and lime at a pH of about 12 resulted in a filtered effluent meeting the above plutonium tolerance.

Treatment Plant Design

The treatment plant was, therefore, designed to provide prolonged flocculation and sedimentation, serial flocculation and sedimentation and filtration through sand at low rates. The flow diagram of the plant is shown in Figure 1. The waste enters the plant over a weir (1) where the pH and flow measurements are made and recorded. Lime and/or caustic soda are added as required for pH control to the flash mixer (2) following which the waste enters the holding tanks (3), (4), (5) and (6). These tanks have a total capacity of 3 days flow and provide hold-up for equalization of waste characteristics. One of these may be used for storing unusual or difficult wastes to treat so that they can be blended gradually with the treatable waste. All of the preliminary storage tanks are of hopper bottom construction for the possible draw off of solids. These tanks are also equipped with stirring mechanisms for holding solids in suspension when that is desired. The neutralized waste is pumped to the flocculators (7) and (9). Here the coagulants or any combinations of the coagulants or absorbents are supplied. The flocculators and the settling tanks following them (8), (10) may be operated in parallel or used for serial treatment. With serial operation the theoretical detention time for average flows is 60 minutes in each flocculator and 6 hours in each settling tank. The flocculator effluent enters the settling tank through a slightly submerged inlet and the supernatant is taken over a weir at the opposite end of the tank to the filters (11), (12), (13). The settling tanks are also hopper bottom for collection and removal of chemical sludge.

The three sand filters contain 18 inches of gravel in 5 layers of graded sizes from 3/16 inches to 1-1/2 inches in diameter. The sand includes two layers consisting of a bottom layer of coarse sand 3 inches in thickness placed directly on top of the gravel and the upper layer of filter sand proper with a depth of 15 inches. The specifications for the coarse sand calls for a grain size ranging from 1 mm. to 3.0 mm. with not less than 2 per cent by weight retained on a No. 8 sieve and not more than 98 per cent by weight retained on a No. 12 sieve. The filter sand proper has an effective size of not less than 0.44 mm. and not more than 0.52 mm. and a uniformity coefficient not to exceed 1.7. To meet this specification, practically all of the sand grains must be finer than 1.0 mm., and not more than 2 per cent can be finer than 0.3 mm. in diameter. For comparative purposes the sand in these filters will be removed at a later date and the media replaced with anthrafilt media with an effective size between 0.55 and 0.65 mm., and a uniformity coefficient not in excess of 1.75. Each filter is equipped with a loss of head gauge of the lever and weight controlled diaphragm type. These filters are equipped with the conventional washing facilities as well as a surface wash system. The under drain system consists of a manifold with 2 inch cast iron pipe laterals spaced on one foot centers. The rate of filtration has been varied between 1 and 3 gallons per square foot per minute. This may be compared to normal rates of 2 to 4 gallons per square foot per minute in normal municipal water works practice. To date no difficulties have been experienced with the filter beds. Experience has indicated that the

plutonium activity is retained with the fine floc on or near the surface of the sand. The filter effluent is collected in effluent holding tanks where it is monitored to assure that it is within the 70 counts per minute limit for plutonium before it is discharged into the canyon. The filters are backwashed with effluent water and the dirty wash water is returned to the primary holding tanks for retreatment. Suspended solids and sludge collection is provided where sedimentation might take place. The settled sludge flows by gravity to a sludge concentration tank, (15), where further concentration by sedimentation takes place. The supernatant from the sludge concentration tank is returned for retreatment. The concentrated sludge is pumped with diaphragm pumps, (16), to the vacuum filter, (17), for dewatering.

The materials of construction are concrete and other materials commonly used in water treatment plants. Stainless steel and duriron are used only on the influent channel where the metals are in contact with the unneutralized wastes.

Operation of Plant

After some difficulties due to faulty operation of certain appurtenances the plant was put into operation on dummy runs with uncontaminated wastes. During this trial the operators were broken in on the proper use of all valves, pumps, tanks, flocculators, sand filters, etc. These trials indicated the need for distribution baffles near the influent end of the sedimentation tanks. Such baffles were installed

and improved operation in sedimentation was obtained. The plant was then put into operation on laboratory wastes on an eight hour 5 day per week basis. This schedule of operation was possible because of the capacity of the storage and equalizing tanks. As the plant is still in the "shake-down" period of operation, more analytical and control work is necessary for developing the best operating techniques and control procedures. At present two engineers are operating this plant and two chemists are required for carrying out all the analytical determinations necessary for its proper operation and control. In addition, the usual part-time supervision is required for analytical control and operation by one additional chemist and one engineer. Because of the frequent changes in waste characteristics and the necessity for determining optimum coagulation doses and conditions for maximum plutonium removal, a plant of this type requires more careful control of operating performance than an ordinary water purification plant. A series of jar coagulation tests must be made regularly to determine the optimum coagulation combination and dose required. At present the quality and settling characteristics of the floc is the best parameter available for judging the probable success of plutonium removal. It has been noted that there is no strict correlation between turbidity removal and activity removal for some alpha emitters. A better parameter of control is required than is given by either pH, coagulant dosage, floc formation or turbidity removal. The determination of plutonium is somewhat combersome and time-consuming. Plutonium residuals and removals are valuable for the record but such data are not available in time to be used for plant control.

Collection of Data

Regular samples are collected from the raw influent waste, the waste storage tanks, the flocculator effluents, the settling tank effluents, the filter effluents, the treated water storage tank, and the sludge concentration tank. Composites representing the daily operation are collected at all of these points except the last. The analytical determinations made on the raw and treated waste samples include total and suspended volatile solids and ash, pH, turbidities, gross alpha counts and plutonium counts. Sodium, magnesium, calcium potassium, alkalinity, nitrate and fluoride determinations are made on the raw and stored waste samples.

Since the operation of the plant has begun the characteristics of the raw waste have been somewhat different than expected. Data collected over a typical one-month period of plant operation, as shown in Table 1, indicate that the total solids have varied between 275 p.p.m. and 8,000 p.p.m. with 40 to 65 per cent ash. Although the pH of the waste seems to average around 6.0 variations from below 2 to as high as 12.0 are not unusual. The suspended solids in the raw waste varied between 2 and 2,500 p.p.m. and the turbidities between 75 and 800. During the first 3 month period the average daily plutonium alpha counts in the raw waste varied from 35 to 20,000 per minute per liter with an average of about 2,000 C/M/L. The chemical coagulant doses varied between 20 to 80 p.p.m. of $FeCl_3$ and between 40 to 600 p.p.m. of $Ca(OH)_2$. The filter

delivered an effluent with a turbidity less than 2 p.p.m. Plutonium removals have been excellent. Filter effluent values of 20 to 30 counts per minute per liter (1.8×10^{-5} to 2.7×10^{-5} uc [redacted] per liter) are common and the poorest effluent so far obtained has been 200 C/M/L (1.8×10^{-4} uc/[redacted]/L). As expected, the plutonium removals are greatest when the coagulation tanks and sedimentation tanks are operated in series. Such operation naturally decreases the capacity of the plant. An increase in filtration rates between 1 and 3 gallons per square foot per minute apparently does not materially decrease the efficiency of the plutonium removals. Filter runs at rates of 2 gallons per square foot per minute average about 20 hours. The wash water requirement has averaged about 2-1/2 per cent of the waste treated.

Chemical sludge production is also somewhat higher than was expected, perhaps due to a higher concentration of total solids in the raw wastes. The chemical sludge containing the plutonium as drawn from the settling tanks has averaged about 1 per cent solids. After concentration in the sludge holding tank this sludge contains 1.7 to 2.2 per cent solids. Upon filtration on an EIMCO vacuum filter with diatomaceous earth as a filter aid, the "hot" sludge cake contains between 22 and 25 per cent solids. This wet sludge cake is put into used oil barrels and is stored in the contaminated storage area. The plant performance has indicated a sludge cake production of about 2 gallons per thousand gallons of waste. This means that about 40-fifty gallon barrels will have to be stored for each million gallons of waste that are treated. A change

from slaked lime to the use of caustic soda for neutralizing the raw waste resulted in the production of a somewhat smaller volume of sludge. This shows that considerable space is still required for storing "hot" sludge. However, this is a volume reduction from the untreated waste of about 500 to one which is of the same or slightly greater order of magnitude than can be expected by evaporation. Some additional reduction in volume might be achieved by sludge drying. Sludge drying, however, is not desirable at present because of the additional toxicity hazards involved in possible dust production associated with drying operations.

Although alum has been tried intermittently for coagulation, difficulty was experienced in maintaining the pH in the range suitable for alum coagulation. Consequently, lime and ferric chloride have been used almost exclusively for coagulation.

Septic sludge has been encountered in the sedimentation tanks due to the activity of microorganisms on the organic matter in the sludge. This difficulty has been encountered in the two plants of this type built to date. At Los Alamos this condition has been eliminated by the frequent withdrawal of sludge and the use of chlorine at the influent of the settling tank.

The decontamination factors obtained in this treatment may vary between slightly less than 10 to as much as 10^3. These values are not impressive when compared to the decontamination factors obtained by high efficiency evaporation processes on wastes of higher activity. However, the decontamination obtained on these low level wastes produces

an effluent of drinking water quality as far as plutonium is concerned. The volume of contaminate residue obtained, which is also important, is equal to or better than evaporation processes.

Costs

An estimate of the cost of this treatment has been made on the basis of operating costs including such items as supervision and labor, chemicals, power, maintenance, overhead and plant investment and depreciation. Investment in plant was written off at the rate of 10 per cent per year. On this basis the overall cost of this treatment is about 0.8 cents per gallon. This compares favorably with costs of evaporation which have been estimated to be as high as 13 cents per gallon. However, this cost estimate of $8,000 per million gallons is a tremendous figure when compared to municipal water treatment at less than $50 per million gallons. The difference may be explained on the basis of volumes of waste to be treated, on initial cost of plant and equipment, and on higher supervision, operation and maintenance costs. About 90 per cent of this cost is for supervision and labor and plant depreciation.

It should be noted that wastes are obtained containing plutonium which cannot be treated by the coagulation process because the concentrations of detergents, citrate, or polyphosphates prevent floc formation. Some laundry wastes belong in the latter category. Such wastes may be treated biologically on two-stage high ratio recirculating trickling filters. Experiments on the successful pilot plant operation

of trickling filters for the removal of plutonium from laundry wastes have been described by Newell et al [6]. The activated sludge process is not satisfactory in this case because of the propensity for foam production with substrates containing surfactants in aeration processes. Because of this and also the greater stability and simplicity of operation, trickling filters are preferable in this case.

It has also been mentioned [1] that a relatively high level process waste containing about 1.0 per cent of citrate is being treated by a modified activated sludge process. Such a waste would be impractical to treat economically by coagulation or carrier precipitation or ion exchange processes. The zoogleal organisms of the activated sludge process are capable of oxidizing the citrate and producing an effluent with a much reduced organic content. The effluent from such a biological treatment can then be treated without difficulty in the coagulation plant described here.

It may be concluded that proper combinations of chemical precipitation and biological processes can be used to treat some refractory wastes containing radioactive materials that could not be treated by either type of process alone. The public and the profession may be assured that progress is being made in the development of new and cheaper methods for the disposal of wastes containing radioactive isotopes in Atomic Energy Commission installations. A number of other Government agencies, notably the U. S. Public Health Service, the U. S. Geological Survey, and the Tennessee Valley Authority, have been cooperating with Commission scientists in the solution of specific liquid waste problems.

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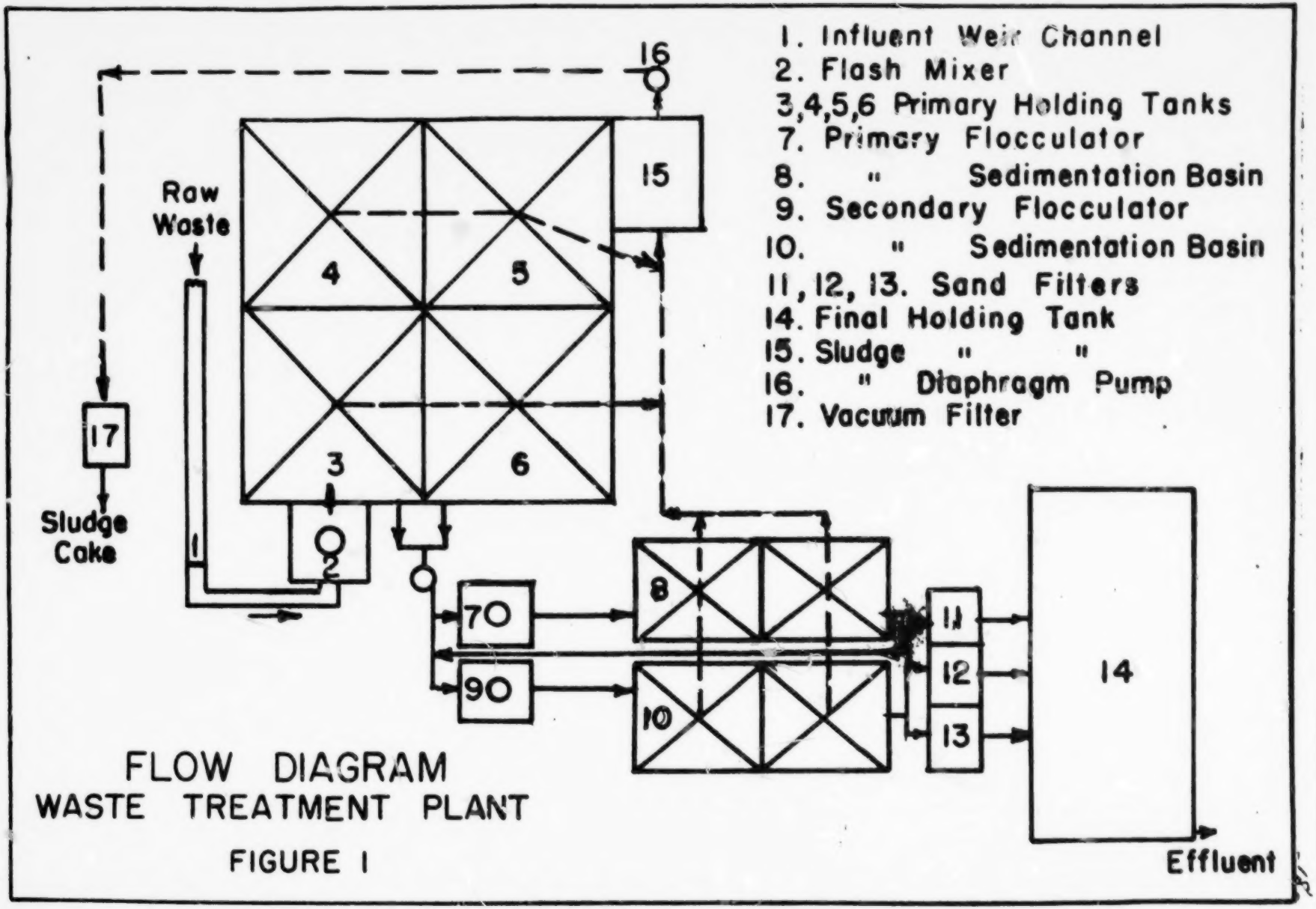
TABLE NO. 1

LABORATORY WASTE TREATMENT PLANT

TYPICAL ANALYTICAL DATA FOR ONE MONTHS OPERATION*

	TOTAL SOLIDS P.P.M.		SUSPENDED SOLIDS P.M.M.		TURBIDITY P.P.M.			PU CONCENTRATION COUNTS PER MINUTE PER LITER		
	Raw Waste	Treated and Filtered Effluent	Raw Waste	Treated Settling Tank Effluent	Raw Waste	Treated Settling Tank Effluents	Treated Filter Effluents	Raw Waste	Treated Settling Tank Effluents	Treated Filtered Effluents
Max. Day	8,000	2,355	2,500	33	300	38	5	5,090	800	153
Min. Day	275	750	2	11	75	4	0.2	35	10	4
Average	1,850	1,590	160	17	365	11	1.5	992	230	25

* The plant was operated under serial treatment for a short period during the month. The maximum plutonium in the filter effluent was obtained during the period of parallel operation.



FLOW DIAGRAM
WASTE TREATMENT PLANT

FIGURE 1